Journal of Magnetism and Magnetic Materials 326 (2013) 151–156

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Electron paramagnetic resonance study of $(La_{0.33}Sm_{0.67})_{0.67}Sr_{0.33-x}Ba_xMnO_3$ ($x<0.1$): Griffiths phase

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ARTICLE INFO

Article history:
Received 18 June 2012
Received in revised form
13 August 2012
Available online 7 September 2012

Keywords:
Doped manganites
Electron paramagnetic resonance
Phase separation
Variable range hopping

ABSTRACT

Manganite compounds $(La_{0.33}Sm_{0.67})_{0.67}Sr_{0.33-x}Ba_xMnO_3$ with light Ba doping ($x=0.01$–0.09) have been investigated by electron paramagnetic resonance over the temperature range 110–450 K. It was found that the EPR linewidth behavior changed drastically in samples with these low Ba concentrations. For all the samples there was observed coexistence of paramagnetic and ferromagnetic phases below the phase-transition (Curie) temperature. EPR signals characteristic of a Griffiths phase were observed in the samples with $x=0.03$, 0.06, and 0.09. The temperature dependence of EPR linewidth in the paramagnetic phase was analyzed on the basis of the variable-range-hopping model, which explained the observed data.

1. Introduction

As reported previously [1–4], the magnetic properties and conductivity behavior of the manganite sample $(La_{0.33}Sm_{0.67})_{0.67}Sr_{0.33-x}Ba_xMnO_3$, with $x=0.0$, containing no Ba^{2+} ions, characterized by the phase-transition temperature $T_C=205$ K, are significantly different from those of the samples containing rather large amount of Ba ions, with $x=0.13$, 0.23, and 0.33, characterized by $T_C=127$ K, 112 K, and 96 K, respectively [1]. This indicates that T_C decreases sharply from 205 to 127 K for a rather small Ba doping of $x=0.13$, and then only to 96 K for Ba doping with the largest possible amount of $x=0.33$. In addition, the resistivity changed from $10^{-1}\,\Omega$ cm for $(La_{0.33}Sm_{0.67})_{0.67}Sr_{0.33}MnO_3$ at 100 K to about $10^3\,\Omega$ cm for all other samples ($x=0.13$, 0.23, 0.33) [1]. Thus, significant differences in magnetic and charge-transfer properties were observed in samples with Ba doping of $(La_{0.33}Sm_{0.67})_{0.67}Sr_{0.33-x}Ba_xMnO_3$. Therefore, investigation of behavior of such samples with light doping of Ba ions ($x<0.10$) is of special interest to develop further insight into the interactions among the various paramagnetic ions in these samples.

This paper reports an investigation of four manganite samples, $(La_{0.33}Sm_{0.67})_{0.67}Sr_{0.33-x}Ba_xMnO_3$, $x=0.01$, 0.03, 0.06, and 0.09, by X-band (~9.45 GHz) electron paramagnetic resonance (EPR) in the temperature range 110–450 K. In particular, the effects of substitution of the Sr^{2+} ion, with the ionic radius of 1.12 Å by the Ba^{2+} ion with the larger ionic radius of 1.34 Å, on the EPR spectra are investigated here. This substitution affects the structure of these compounds leading to a deviation from the structure of stoichiometric $LaMnO_3$, which is orthorhombic with space group Pnma ($T_N=140$ K) [5]. The degree of deviation depends on the ionic radius of the substituting divalent cation, leading to the rhombohedral structure with space group $R\bar{3}c$ for both $La_{0.67}Ba_{0.33}MnO_3$ ($T_C=343$ K) [6,7] and $La_{0.67}Sr_{0.33}MnO_3$ ($T_C=376$ K) [8,9].

2. Effect of ionic substitution in manganites

The amount of substituting ions governs the fractional contents of the Mn^{3+}($3d^4$, $L=2$) and Mn^{4+} ($3d^3$, $L=3$) ions present in the stoichiometric samples with the common formula $La_{0.67}B_{0.33}(Mn^{3+})_{0.67}(Mn^{4+})_{0.33}O_3$. The change in the average size of the cations at the B site of these perovskites causes large changes in their transport and magnetic properties due to modification of the Mn–O–Mn bond angles and Mn–O distances, thereby influencing the e_g electron hopping between the Mn^{3+} and Mn^{4+} states. In turn, such distortion of the bond angles and distances gives rise to competing superexchange and double-exchange interactions, causing electronic phase separation. Furthermore, increasing the Ba content in mixed Sr–Ba manganite leads to increasing disorder in the system, associated with an increasing spin-glass like behavior in these samples. Since diamagnetic La ions are partly substituted by the paramagnetic Kramers Sm^{3+} ions ($J=5/2$, $L=5$, $\mu_{eff}=1.5$) in these compounds, their magnetic states are strongly affected below their phase

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http://dx.doi.org/10.1016/j.jmmm.2012.08.017